

# A quantum critical point in the transverse field of $\text{Mn}_{12}$ system

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## Abstract

Using exact diagonalization method, we studied the ground state of the anisotropic molecular magnets and find a critical point in the transverse field, which may divide the quantum tunneling region into two different parts. Possible ways to observe and take advantage of this point by varying the transverse field are suggested.

Recently the macroscopic quantum phenomena in large spin anisotropic magnets is attracting much attention among both theorists and experimentalists[1-2]. Many efforts have been done to measure the step-wise magnetization loop, classify different temperature regimes, and find different theoretical explanation for each. Quantum resonant tunneling is the underlying mechanism that leads to abrupt change in the magnetisation. However, it's not easy to take into account various factors that may influence the result quantitatively, which sometimes results in sheerly different pictures[3-6]. Fortunately, the model Hamiltonian has been established and widely used in theoretical analysis. Many works have been starting from the single large spin model, treating other parts as mean fields, with some static or dynamic distribution function. There are three different temperature regimes that can be described as different phases: thermal-activated regime, thermal-assisted regime, and purely quantum tunneling regime. Apparent phase transition (PT) has been studied by analytical and numerical means and both first and second order phase transition are found[7]. In this paper, through a properly defined order parameter, we find a kind of quantum phase transition (QPT) for  $Mn_{12}$  which is induced by continuous change of transverse field in the third regime. Its experimental consequences are suggested.

Firstly, the model Hamiltonian of  $Mn_{12}$  is

$$H = -D(S^z)^2 - B(S^z)^4 - C((S^+)^4 + (S^-)^4) - g\mu_B h_x S^x - g\mu_B h_z S^z \quad (1)$$

where the first and second terms are diagonal part of the crystal field, the third term is the off-diagonal part. The last two terms represent external field. In writing down it,

we neglect all the other internal degrees of freedom of the cluster on one lattice site, i.e., the high energy spectrum which can be deviating the description of fixed spin magnitude, for the antiferromagnetic coupling between internal spins (4 spins 3/2 and 8 spins 2 for  $\text{Mn}_{12}$ ) is rather large. This is called the “mesoscopic” approach. The parameters

are determined by various measurements. In the following we choose  $D \approx 0.60K$ ,  $B/D \approx 0.002$ ,  $C \approx 3 \cdot 10^{-5}K$ , which are obtained in EPR experiments. Furthermore we set  $h_z = 0$  to study pure transverse field effect.

In the pure quantum relaxation regime, without the longitudinal field, tunneling occurs between degenerate ground states  $-S$  and  $+S$ . In the Landau-Zener theory the transition probability is given by:

$$P_{-S,S} = 1 - \exp(-\Gamma_{-S,S}) \quad (2)$$

with

$$\Gamma_{-S,S} = \pi \Delta_{(-S,S)}^2 / [4\hbar S g \mu_B dH/dt] \quad (3)$$

which is determined by tunneling splitting as well as sweeping rate of applying field. In this paper, however, we will study a related problem—the characteristic of the ground state—and understand the physics in a different way. We consider the states with largest  $|\langle S_z \rangle|$ , denoted as  $|g\rangle$ , which is also energetically lowest in most cases. Generally  $|g\rangle$  can be expanded as

$$|g\rangle = \sum_{m=-S}^S c_m |m\rangle \quad (4)$$

Now we define order parameter as the difference of probability of the  $|\pm S\rangle$  components in  $|g\rangle$ :

$$\delta = |c_S| - |c_{-S}| \quad (5)$$

For situations where quantum tunneling due to ground state dynamics dominates in sufficiently low temperature,  $\delta$  measures the ability of ground state  $|g\rangle$  to transfer spins

from one side in magnetic spectrum to other side. Some cases corresponding to (1)  $B = 0, C = 0$ . (2)  $B = 0.002, C = 0$ . (3)  $B = 0, C = 10^{-5}$ , are studied and demonstrated in Figure 1. The  $\delta$  line changes abruptly near some point of  $h_x$ , showing a quantum critical point  $h_c$  in the classical limit  $S \rightarrow \infty$ . Across this narrow crossover region, the difference of  $|\pm S\rangle$  components in  $|g\rangle$  (i.e.,  $\delta$ ), changes from almost 1 to 0. When  $|h_x| > h_c$ , numerical results show equal components of the “bare” states  $S = \pm 10$ , in our case. The effect of  $B$  and  $C$  term on the  $\delta$  line, especially on the crossover position are easy to understand.  $B$  term tends to strengthen the anisotropy, leading to higher barrier between the positive and negative half of the magnetic spectrum. Thus a larger value of  $|h_x|$  is needed to cause tunneling combination. However, the  $C$  term is transverse and will help tunneling, and because of its four-ordered operator form, can influence the results greatly by a little change of its value ( $10^{-5}$  relative to  $D$ , and  $10^{-2}$  to  $B$ ). The combined effect is shown in Figure 2. The  $C$  term quickly decreases the difference of the two “phases”. In some systems,  $C \approx 3 \cdot 10^{-5}$  so maybe we need some trick to really observe the critical point.

Also we calculated the distribution on bare states of  $|g\rangle$ . As is evident from Figure 3. The  $|\pm S\rangle$  components are sizable near the crossover region in the  $\delta = 0$  phase. The experimental consequence is, in a well prepared sample, if we drive all sample sites to  $|S = 10\rangle$  state, a considerable portion of lattice site can tunnel to the  $|S = -10\rangle$  state through the ground state channel, if the transverse field is properly controlled. Or, by increasing the transverse field across the crossover region, we can “pump” many sites from one side of the magnetic spectrum to another. Such scenario is related to relaxation experiment [8].

To conclude, by means of exact diagonalization, we find a crossover in the Hamiltonian of molecular magnets, through which the difference of amplitudes of  $|\pm S = 10\rangle$  states in ground state are changed from almost 1 to zero. This property might have some experimental consequences in the purely quantum regime of the magnetization relaxation which is caused by tunneling through the ground state channel.

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## Caption:

Figure 1. The amplitude difference  $\delta$  of the  $|\pm S\rangle$  components in the ground state  $|g\rangle$  for three groups of  $B, C$  values. The  $\delta$  line changes abruptly near some  $h_x$  points. The effect of  $B$  is to broaden the central “phase” of the diagram, while the  $C$  value effects the opposite way.

Figure 2. For larger  $C$  values the central phase narrows quickly and will disappear at large enough  $C$  value. This made observation of the critical point of transverse field more subtle and difficult. Here we plotted two lines corresponding to  $C = 10^{-5}$  and  $C = 2 \cdot 10^{-5}$ .

Figure 3. The amplitudes of the ground state projected to bare states:  $|-S\rangle, \dots, |S\rangle$ , for cases (a)  $h_x = 1$ , (b)  $h_x = 2.3$ , (c)  $h_x = 2.7$ , (d)  $h_x = 3.3$ . Both  $B$  and  $C$  are chosen to be zero. The distribution tends to become symmetric as  $h_x$  transcends some value.

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